Investigation of the Flow Boiling Heat Transfer Characteristic for Non-Flammable Cryogenic Mixed Refrigerant

J. Yoo, L. Jin, C. Lee, S. Jeong

Cryogenic Engineering Laboratory, Korea Advanced Institute of Science and Technology, Daejeon, 34141, South Korea

ABSTRACT

An experimental apparatus is fabricated to measure the flow boiling heat transfer coefficients (HTC) of cryogenic mixed refrigerants (MR). The experimental system has a test section which includes a copper tube and a thin-wire heater to impose a heat flux on the experimental MR stream. The flow boiling heat transfer characteristics (HTC) are investigated for three cases of non-flammable cryogenic MR sets. R218 (Octafluoropropane, C_3F_8) and R14 (Tetrafluoromethane, CF_4) are used to constitute the experimental MRs. Three different molar compositions are selected to obtain the experimental HTC of the non-flammable mixtures (i.e. R218:R14 = 0.7:0.3 / 0.5:0.5 / 0.3:0.7). The mass flux conditions are varied as 88, 198 and 330 kg/m²s during the experiment. The heat flux conditions are selected as 2800 and 9900 W/m². Experimental measurements are conducted for the whole MR sets at the pre-determined physical conditions. The obtained heat transfer characteristic results have 19.1% of mean uncertainty. Also, we compare three correlation sets to identify the best correlation which estimates the experimental results with the good accuracy. The most accurate correlation is Kandlikar's correlation with Fujita and Tsutsui's mixture correction factor. It predicts the experimental data within 28.2% in mean deviation.

INTRODUCTION

Recently, many efforts have been made to utilize non-flammable mixed refrigerant (MR) rather than hydrocarbon as the working fluid of an MR Joule-Thomson (JT) refrigerator because of the safety issues [1-3]. R218 (Octafluoropropane, C₃F₈) and R14 (Tetrafluoromethane, CF₄) have fairly good thermodynamic properties to be utilized as the working fluids for refrigerators from 300 K to 145 K. For this reason, R218 and R14 can be good non-flammable substitutes for propane and methane. When a MR JT refrigerator is constructed, the heat exchanger is considered as an important component which affects the size and the cost of the refrigerator. A efficient and compact heat exchanger is required to implement an excellent MR JT refrigerator. The precise estimation the heat transfer area needed is important to design of a compact heat exchanger. Since the heat exchange process in the MR JT refrigerator occurs in the form of evaporation through boiling and condensation, the flow boiling heat transfer characteristics (HTC) of the working fluid are key factors for determining the precise heat transfer area. Since little research has been conducted on the HTC of non-flammable MR, it is hard to determine the exact heat transfer area of the heat exchanger for

non-flammable MR [4]. Understanding the flow boiling HTC for a specific non-flammable MR, is necessary before it is used as the working fluid of the MR JT refrigerator. Since it is difficult to identify all of the necessary flow boiling HTC by experiments, a correlation for estimating the flow boiling HTC is suggested with the acquired HTC information for the non-flammable MR.

In this paper, we examine the ensuring trustworthiness of the experimental apparatus as well as the experimental device for measuring the flow boiling HTC of a R218 and R14 mixture. The experimentally obtained HTC results are presented with their measuring uncertainties. With the experimental data, three correlations are compared to examine which correlation can describe the experimental data with minimum error. Two of the three correlations have already been suggested by previous researchers, Thome and Barraza [4, 5]. Another new one is proposed by us.

EXPERIMENTAL APPARATUS

Overall system

The schematic diagram of the overall experimental system is shown in Figure 1. In the experimental system, a refrigerant circulation system is required to create the flow of the working fluid. A compressor functions with the predetermined experimental pressure and mass flow conditions as the main component of the circulation system. The compressed working fluid, which reaches approximately 373 K, is chilled by water at the after-cooler. A throttling valve and a mass flow meter are installed next to the after-cooler. The throttling valve and the mass flow meter are used to adjust and record the mass flow rate of the working fluid, respectively. After passing through the mass flow meter, the stream is fed into a cold box.

The cold box is maintained at high vacuum condition (below 1.3×10^{-2} Pa) to avoid heat penetration from the environment. In the cold box, the working fluid is cooled down completely to the bubble point by a Stirling refrigerator (CryoTel-GT, Sunpower). The trim heater (customized-KHLV, Omega) is located at the exit stream of the refrigerator heat exchanger. It supplies heat to the liquid state refrigerant stream to precisely control the experimental temperature conditions. Then, the stream which has been finally characterized as the specific experimental temperature, pressure and mass flux condition enters the test section.

Test section

The test section is illustrated in Figure 2. The test section contains a horizontal copper tube which is 1.7 mm in the inner diameter, 3.2 mm in the outer diameter and 100 mm in length. The thin manganin wire heater (MW-32, Lakeshore) wraps the copper tube to induce heat exchange

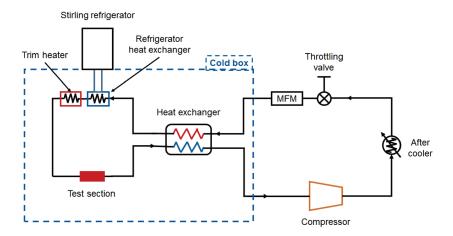


Figure 1. Schematic diagram of the overall experimental system

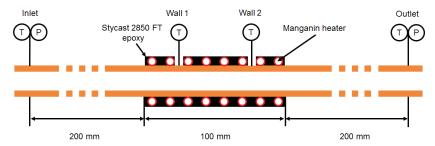


Figure 2. Schematic diagram of the test section

process between the heated wall of the test section and the MR stream through the test section. Epoxy (Stycast 2850 FT) is used to improve the thermal contact between the heater and the copper tube. The test section is instrumented by several temperature and pressure sensors. There are four temperature sensors (SD-670, Lakeshore) and two pressure sensors (FP2000, Honeywell) in the test section. The temperature sensors are attached on the inlet (T_{in}), the outlet (T_{out}) and the outer-wall of the test section ($T_{wall,I}$, $T_{wall,2}$), respectively. The pressure sensors are installed on the inlet (P_{in}) and the outlet (P_{out}) of the test section. The flow boiling HTC for the experimental MR sets are to be obtained by logarithmic mean temperature difference (LMTD) method with the collected temperature and pressure data. Equations for LMTD method are as follows [6]. All of the necessary thermodynamic properties of mixture for obtaining experimental HTC are calculated by REFPROP 9.1 [7].

$$\Delta T_{wall,in} = T_{wall} - T_{in} \tag{1}$$

$$\Delta T_{wall.out} = T_{wall} - T_{out} \tag{2}$$

$$\Delta T_{LMTD} = \frac{\Delta T_{wall,in} - \Delta T_{wall,out}}{ln(\frac{\Delta T_{wall,in}}{\Delta T_{vall,vot}})}$$
(3)

$$A_{s} = \pi D_{i}L \tag{4}$$

$$h_{exp} = \frac{q''}{A_s \Delta T_{LMTD}} \tag{5}$$

where T_{in} , T_{out} and T_{wall} indicate the inlet, outlet and the wall temperature of the test section, respectively. Also, A_s is the heat transfer area between the heated test section surface and the working fluid stream. D_i is the inner-diameter of the test section and L is the length of the test section. In Equation (5), h_{exp} represents the experimentally obtained flow boiling HTC value. q " is the imposed heat flux to the test section.

Calibration of the experimental apparatus

The measurement accuracy of the experimental apparatus should be confirmed to measure the unknown HTC of refrigerants such as a mixture of R218 and R14. In this paper, the experimental apparatus is calibrated with a gas phase nitrogen which has well-known HTC estimation results from the Dittus-Boelter's correlation. It is described by Equation 6 [6].

$$Nu_D = 0.023Re_D^{0.8}Pr^{0.4} (6)$$

where Nu_D , Re_D and Pr are the Nusselt number, the Reynolds number and the Prandtl number, respectively.

The main issue for the calibration process is to determine the actual heat transfer area between the hot inner wall of the test section and the fluid in the test section. Due to the axial conduction heat transfer on the test section surface, the equivalent heat transfer length is extended during the heat exchange process. As shown in Figure 3, the actual heat transfer area should be larger than the measured area. It directly affects the experimentally obtained HTC values from Equation 5. For this reason, the actual heat transfer area should be considered to get the accurate flow boiling HTC with LMTD. In this process, the 'equivalent length' concept is applied to reflect the effect of the extended area. The equivalent length is the increased length of the test section, which can explain the effect of the extended heat transfer area. Figure 3 illustrates the 'equivalent length concept by a schematic diagram.

In the calibration process, the estimated HTC results with the Dittus-Boelter's correlation are considered as the true values of the nitrogen gas' HTC. First, the experimentally obtained HTC of the nitrogen gas is compared with the theoretically expected HTC results. For the next step, the equivalent length is deduced by minimizing the errors between the experimental and the theoretical results. Figure 4 (a) shows the experimental results with the apparent length of 100 mm. They have $+5\sim+20\%$ errors compared to the calculation results. Figure 4 (b) represents the comparison results with 110 mm in the equivalent length. In this case, the errors are minimized as $-3\sim+10\%$. As a result, we decide 110 mm as the true equivalent length for subsequent experiments.

EXPERIMENTAL RESULTS

The whole set of experimental conditions is summarized in Table 1. There are three mass flux conditions, 88, 198 and 330 kg/m²s. The heat flux conditions are varied from 2800 to 9900 W/m².

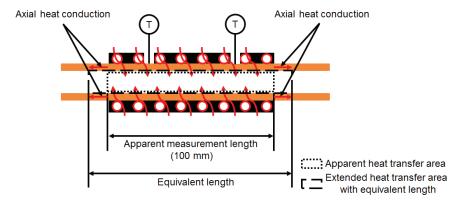


Figure 3. Picture for the equivalent length concept

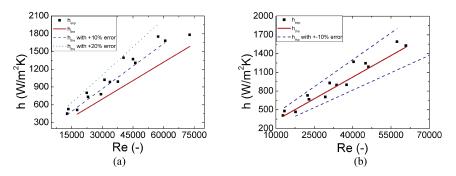


Figure 4. Comparison results between the experimental and estimated HTC results for gas phase nitrogen with (a) 100 mm in the equivalent length and (b) 110 mm in the equivalent length.

Case	Molar composition (-)	Pressure (kPa)	Heat flux (W/m²)	Mass flux (kg/m²s)
1 2 3 4 5 6	R218:R14 = 0.7:0.3	450 ± 150	2800 2800 2800 9900 9900 9900	88.0 198 330 88.0 198 330
7 8 9 10 11 12	R218:R14 = 0.5:0.5		2800 2800 2800 9900 9900 9900	88.0 198 330 88.0 198 330
13 14 15 16 17	R218:R14 = 0.3:0.7		2800 2800 2800 9900 9900	88.0 198 330 88.0 198 330

Table 1 Experimental conditions for measuring R218 and R14 mixtures

The experiments are conducted for three mixtures of MR molar compositions (i.e. R218:R14 = 0.7:0.3 / 0.5:0.5 / 0.3:0.7). A total of 200 data points are experimentally obtained. The whole data sets are plotted in Figure 5. There are measurement uncertainties caused by the resolution of the temperature sensors and the measurement of the actual heat transfer area. The uncertainty of each HTC data set is analyzed by a general method [8, 9]. The uncertainties are marked in each graph by a y-axis error bar. The mean uncertainty of the experimental HTC results is 19.1%. In these experiments, the uncertainties of quality are not denoted in Figure 5 because the quality differences between the inlet and the outlet of the test section are less than 5% for the whole experimental conditions.

According to the experimental results, the flow boiling HTC of the mixtures which are composed of R218 and R14 are distributed in the range of 500 to 5000 W/m²K. The peak HTC values exist at a quality between 0.7 and 0.9. After the maximum point, the HTC drastically decreases because of the dry-out of the residual liquid. The results generally show high flow boiling HTC values at high mass and heat flux conditions.

CORRELATION SELECTION FOR EXPERIMENTAL MR

The HTC correlation which well represents the experimental results should be formulated by using flow boiling HTC of the experimental mixture for the non-experimental conditions. There are two suggested correlations by previous researchers [4, 5]. They estimate the MR flow boiling HTC with high accuracy in the reference papers. Also, we find a fairly good correlation for our experimental results. In this paper, three different correlations are compared to find the most appropriate correlation for our experimental results. The mean deviation is used as the criterion for correlation accuracy. It means the absolute difference between the estimated and the measured HTC values as written in Equation 7. In Equation 7, N indicates the number of experimental data.

Mean deviation =
$$\frac{1}{N} \sum_{i=1}^{i=N} |[h_{the} - h_{exp}] / h_{exp}| \times 100$$
 (7)

Correlations review

There are two types of correlations to be useful for mixture's flow boiling HTC. The first type is originally developed for single-component fluid with a mixture correction factor as defined by Kandlikar [5]. The second type of the correlation is only for mixtures as defined by Little [4].

The first type correlation generally consists of addition of two terms which are convective and nucleate boiling HTC terms [5]. In order to apply the single-component correlation to the mixture, all of the required thermodynamic properties should be substituted for those of the mixtures. In addition, the mixture correction factor needs to be multiplied by the nucleate boiling term because

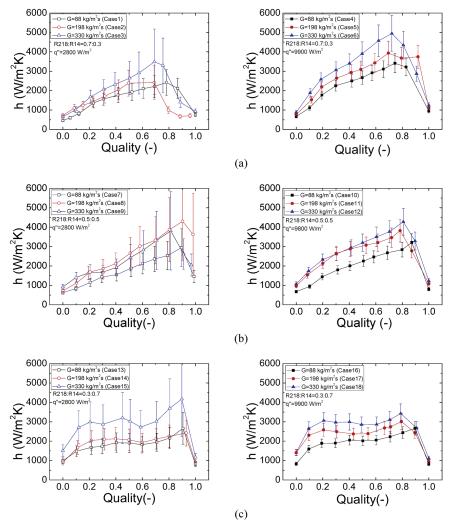


Figure 5. Experimental results for (a) (R218:R14=0.7:0.3) mixture, (b) (R218:R14=0.5:0.5) mixture and (c) (R218:R14=0.3:0.7) mixture

the mass diffusion effect in mixtures degrades the HTC of the mixtures [10-15]. Thome suggests Gungor and Winterton's correlation with Thome's mixture correction factor for three mixture, R402A (R125:R290:R22 = 0.51:0.04:0.45, by mole fraction), R404A (R125: R143A:R134A = 0.36:0.60:0.04, by mole fraction) and R502 (R22:R115 = 0.63:0.37, by mole fraction). According to the reference paper, the suggested correlation has 27.8% in the mean deviation [5, 16]. The detailed correlation and the mixture correction factor are expressed as follows;

$$h_{tp} = E h_L + S h_{nb} \times \frac{1}{1 + \phi} \tag{8}$$

$$\phi = \frac{\Delta T_{bp}}{\Delta T_{id}} \left[1 - exp(\frac{-Bq''}{\beta_l \rho_l i_{fg}}) \right]$$
(9)

where h means the HTC and ϕ indicates the mixture correction factor. The subscript tp, L and nb represent two-phase, liquid phase and nucleate boiling state, respectively.

In this paper, another first type correlation is also examined to find an accurate correlation for the experimental data. Kandlikar's correlation with Fujita and Tsutsui's mixture correction factor is adopted for this purpose. This combination of the correlation and the mixture correction factor is newly suggested in this paper. The correlation with the suggested mixture correction factor is shown in Equation 10 and Equation 11. In Equation 10, the first term at the right side signifies the convective heat transfer term and the second term represents the nucleate boiling heat transfer term. There is an fluid dependent parameter, F_{pp} in Kandlikar's correlation. The parameter accounts for the characteristic of the fluid. It can be determined by experimental data. Table 2 shows the fluid dependent parameters of some refrigerants [17]. As shown in Table 2, the parameters for the low boiling point fluids such as nitrogen and neon are higher than those of high boiling point fluids. The normal boiling point of R218 and R14 are 236 K and 145 K, which are relatively high boiling points compared to nitrogen and neon. The average of F_{pl} values for the high boiling point fluid (i.e. F_{pl} values of water, R11, R12, R13B1, R22, R113, R114 and R152a) in Table 2, therefore, is used as the fluid dependent parameter of the experimental mixture. For this reason, the average value 1.4 is firstly considered as the fluid dependent parameter for the experimental mixtures of R14 and R218.

$$h_{tp} = C_1 C o^{C_2} h_L + C_3 B o^{C_4} F_{fl} h_L \times \frac{1}{1 + \phi}$$
 (10)

$$\phi = \frac{\Delta T_{bp}}{\Delta T_{id}} \left[1 - exp \left\{ \frac{-60q''}{\rho_v i_{fg}} \left(\frac{\rho_v^2}{\tau g(\rho_l - \rho_v)} \right)^{1/4} \right\} \right]$$
 (11)

where Co is convection number and Bo is boiling number. F_{fl} means fluid dependent parameter which reflects the effect of each fluid.

As the second type of the correlation, in this paper, the correlation which is developed by Barraza et al., is selected. Barraza et al., modified Little's correlation to improve its accuracy for newly tested seven kinds of mixtures [18]. Their experimental mixtures include both hydrocarbon and non-flammable. The mean deviation of the suggested correlation is 18% [4]. The modified Little's correlation appeared as Equations 12 through 15.

$$\frac{1}{h_{lp}} = \frac{1}{h_{l,film}} + \frac{x^2 C p_v^2}{\left(\left(1 - x\right) C p_l + x C p_v\right) \left(\frac{\partial i}{\partial T}\right)_p} \frac{1}{h_{lv}}$$
(12)

$$h_{l,film} = 0.023 \left(\frac{\text{Re}_l}{1 + \sqrt{\alpha}}\right)^{0.8} \text{Pr}_l^{0.4} \frac{k_l}{ID(1 - \sqrt{\alpha})}$$
 (13)

$$h_{lv} = 0.023 \left(\frac{\text{Re}_{v}}{\sqrt{\alpha}}\right)^{0.8} \text{Pr}_{v}^{0.4} \frac{k_{v}}{ID\sqrt{\alpha}}$$
(14)

1	1 1	
Fluid	Normal boiling point	Fluid dependent parameter(F_n)
(-)	(K)	(-)
Water (H ₂ O)	373	1.00
R11 (CCl ₃ F)	297	1.30
R12 (CCl ₂ F ₂)	243	1.50
R13B1 (CBrF ₃)	215	1.31
R22 (CHClF ₂)	232	2.20
R113 (CCl ₂ FCClF ₂)	321	1.30
R114 (CCIF, CCIF,)	277	1.24
R152a (CHF,CH ₃)	249	1.10
Nitrogen (N ₂)	77.6	4.70
Neon (Ne)	27.1	3.50

Table 2 Examples of the fluid dependent parameters for various fluids

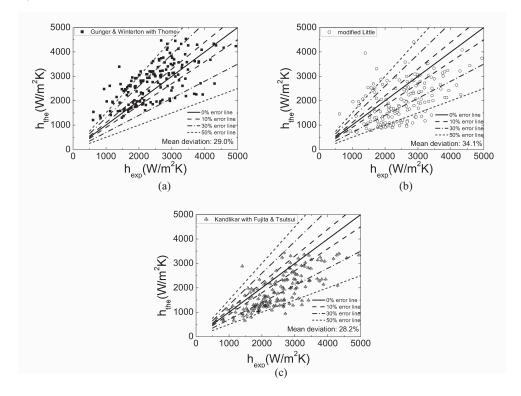


Figure 6. Comparison results of (a) Gungor and Winterton's correlation with Thome's mixture correction factor (b) modified Little's correlation (c) Kandlikar's correlation with Fujita and Tsutsui's mixture

$$\alpha = \left(1 + \frac{\left(1 - x\right)\rho_{\nu}}{x\rho_{l}}\sqrt{1 - x\left(1 - \frac{\rho_{l}}{\rho_{\nu}}\right)}\right)^{-1}$$
(15)

Comparison results for three correlations

All the thermodynamic properties to calculate the correlations are obtained from REFPROP 9.1[7]. The correlations are tested in the quality range from 0.1 to 0.9 of the experimental data. Theoretical HTC values (h_{the}) are calculated by each correlation and are compared with the experimentally obtained HTC (h_{exp}) in Figure 6. As shown in Figure 6(c), Kandlikar's correlation with Fujita and Tsutsui's mixture correction factor has the best accuracy of 28.2% in mean deviation. The mean deviation of Gungor and Winterton's correlation with Thome's mixture correction factor is 29.0% and that of the modified Little's correlation is 34.1%.

The fluid dependent parameter is considered as a correction factor which represents the effect of each single-component fluid. The mixtures of the same components but with different molar compositions can be treated as different fluids. The fluid dependent parameters, therefore, should be varied to the molar composition of the mixture. Although the constant value of 1.4 is used in this paper for the fluid dependent parameter of the experimental mixtures, the Kandlikar's correlation with the Fujita and Tsutsui's mixture correction factor gives the best estimation result. It means that this correlation can be improved if proper fluid dependent parameters are identified for each molar composition of the experimental mixture.

CONCLUSION

This paper describes an experimental apparatus for measuring the flow boiling HTC of the MRs which consist of R218 and R14. The equivalent length concept of the heated area is used to interpret the experimental data because the heat transfer area is very sensitive parameter to obtain experimental flow boiling HTC. The experimental apparatus is calibrated by single phase nitrogen gas.

The experimental flow boiling HTC of the R218 and R14 mixtures are measured from 500 W/m²K to 5000 W/m²K. The mean uncertainty of the experimental results is 19.1%. Three correlations are considered to find the one with the best accuracy. For the comparison results, Kandlikar's correlation with Fujita and Tsutsui's mixture correction factor has the smallest mean deviation of 28.2%. It could be improved to become a more precise correlation when the fluid dependent parameter is appropriately defined for each molar composition of the experimental MR.

ACKNOWLEDGMENT

This research is supported by a grant from Space Core Technology Development Program of National Research Foundation of Korea (NRF-2013-042033) funded by Ministry of Science, ICT & Future planning (MSIP)

REFERENCES

- Walimbe, N.S., K.G. Narayankhedkar, and M.D. Atrey, "Experimental investigation on mixed refrigerant Joule-Thomson cryocooler with flammable and non-flammable refrigerant mixtures," *Cryogenics*, vol. 50, no. 10 (2010), pp. 653-659.
- Boiarski, M., O. Podtcherniaev, and K. Flynn, "Comparative Performance of Throttle Cycle Cryotiger Coolers Operating with Different Mixed Refrigerants," Cryocoolers 13, Springer US, (2005), pp. 481-488.
- 3. Lee, C., et al., "Visualization of the solid–liquid equilibria for non-flammable mixed refrigerants," *Cryogenics*, vol. 75, no. (2016), pp. 26-34.
- 4. Barraza, R., et al., "Description and validation of the Little correlation for boiling zeotropic mixtures in horizontal tubes from cryogenic to room temperature," *IOP Conference Series: Materials Science and Engineering*. IOP Publishing, (2015), pp. 012132.
- 5. Thome, J.R., "Boiling of new refrigerants: a state-of-the-art review," *International Journal of Refrigeration*, vol. 19, no. 7 (1996), pp. 435-457.
- Bergman, T.L., F.P. Incropera, and A.S. Lavine, Fundamentals of Heat and Mass Transfer, John Wiley & Sons, (2011), pp. 578-593.
- 7. Lemmon, E., M. Huber, and M. McLinden, "NIST Standard ReferenceDatabase 23: Reference Fluid Thermodynamic and Transport Properties-REFPROP. 9.1," vol. no. (2010), pp.
- 8. Van Dresar, N.T., "An uncertainty analysis of the PVT gauging method applied to sub-critical cryogenic propellant tanks," *Cryogenics*, vol. 44, no. 6 (2004), pp. 515-523.
- 9. Nellis, G., C. Hughes, and J. Pfotenhauer, "Heat transfer coefficient measurements for mixed gas working fluids at cryogenic temperatures," *Cryogenics*, vol. 45, no. 8 (2005), pp. 546-556.
- 10. Jungnickel, H., P. Wassilew, and W. Kraus, "Investigations on the heat transfer of boiling binary refrigerant mixtures," *International Journal of Refrigeration*, vol. 3, no. 3 (1980), pp. 129-133.
- 11. Thome, J.R., "Prediction of binary mixture boiling heat transfer coefficients using only phase equilibrium data," *International Journal of Heat and Mass Transfer*, vol. 26, no. 7 (1983), pp. 965-974.
- 12. Fujita, Y. and M. Tsutsui, "Heat transfer in nucleate pool boiling of binary mixtures," *International Journal of Heat and Mass Transfer*, vol. 37, no. (1994), pp. 291-302.
- 13. Fujita, Y. and M. Tsutsui, "Heat Transfer in Nucleate Boiling of Binary Mixtures. (Development of a Heat Transfer Correlation)," *JSME International Journal Series B*, vol. 40, no. 1 (1997), pp. 134-141.
- Inoue, T., N. Kawae, and M. Monde, "Characteristics of heat transfer coefficient during nucleate pool boiling of binary mixtures," *Heat and Mass Transfer*, vol. 33, no. 4 (1998), pp. 337-344.

- 15. Sun, Z., et al., "Nucleate pool boiling heat transfer coefficients of pure HFC134a, HC290, HC600a and their binary and ternary mixtures," *International Journal of Heat and Mass Transfer*, vol. 50, no. 1 (2007), pp. 94-104.
- 16. Gungor, K.E. and R. Winterton, "A general correlation for flow boiling in tubes and annuli," *International Journal of Heat and Mass Transfer*, vol. 29, no. 3 (1986), pp. 351-358.
- 17. Kandlikar, S.G., "A general correlation for saturated two-phase flow boiling heat transfer inside horizontal and vertical tubes," *Journal of Heat Transfer*, vol. 112, no. 1 (1990), pp. 219-228.
- 18. Little, W., "Heat transfer efficiency of Kleemenko cycle heat exchangers," *Adv. in Cryogenic Engineering*, Vol. 53, Amer. Institute of Physics, Melville, NY (2008), pp. 606-613.